Empirical model approach for the evaluation of pH and conductivity on pollutant diffusion in soil environment

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A R T I C L E   I N F O

A B S T R A C T

This work is aimed at developing a mathematical model equation that can be used to predict the fate of contaminant in the soil environment. The mathematical model was developed based on the fundamental laws of conservation and the equation of continuity given as $\frac{\partial C}{\partial t} = \nabla.(D\nabla C) - \nabla.(vC) - \frac{\partial Ca}{\partial t} - f$ and was resolved to obtain a quadratic equation of the form $C(X) = DX^2 + vX + f$. The developed equation was then used to fit the experimental data that were obtained from the Physio-chemical analysis of the soil samples which were obtained at various depths; within the vicinity of the H & H Asphalt plant Company, located at Enito 3 in Ahoada West L.G.A, River State, Nigeria. The Experimental and Model results obtained from the Calculation and Simulation of the developed models were compared numerically and graphically as presented in this work. It was observed that there is reasonable level of agreement between the three results. The polynomial of the curve was established to ascertain the validity of the model; this was done for all the parameters that were analyzed. From the findings the model developed can be used to predict the concentration of a chemical pollutant at various depths. The reliability of the model developed was established giving the fact that through this quadratic equation the diffusivity (coefficient of diffusion), the water velocity and the irreversible reaction decay rate could be determined.

Capsule Summary: Mathematical model was developed and used for the prediction of fate of contaminant in the soil environment.

INTRODUCTION

Soil pollution or contamination is caused by the presence of Xenobiotic (human made) chemicals and other alteration in the natural soil environment (Yong, 1992). Waste produced from nature itself such as dead plants, carcasses of animals and rotten fruits and vegetation only adds to the fertility of the soil. However, our waste products are full of chemicals that are not originally found in nature and lead to the soil contamination and pollution (Thomas, 2007). It is typically as result of industrial activities, Agricultural chemical usage and improper disposal of wastes. The most common chemicals involved are petroleum hydrocarbons, pesticides, fertilizers, lead, arsenic, zinc, chromium and other heavy metals. Soil contamination is correlated with the degree of industrialization and the intensity of agrochemical usage (Stagnitti et al., 2003). The study of the degree of deposition

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and diffusion of pollutants in soil is very key and important for a number of reasons. Some chemicals are important as they are required for soil and plant nutrient, other chemicals may be highly toxic particularly if they are present in high concentration. A chemical becomes a pollutant if its concentration exceeds some prescribed water and or soil required quantity/quality standard or if a beneficial use has been impaired, and if the cause is induced by human activities (Parlange et al., 1995).

The investigation of pollutant diffusion in, and through the soil environment majorly centers on this zone. Mathematical models are developed based on the flow mechanism in this zone (Jirka et al., 2002).

The geological media between the land surface and the regional water table below is known as the vadose zone (Stephens, 1996). The zone includes the crop root layer; it therefore, plays an integral role in the global hydrological circle controlling surface water infiltration, runoff, Hence, the availability of soil water and nutrient for plant (Ukpaka, 2014a – i; Amadi and Ukpaka, 2014a – d; Ikenyiri et al., 2012, 2012a).

Initially, investigation of this zone was focused on water availability for crops and optimal management of the root zone. However, in recent years much of increased use of agrochemicals such as Pesticides, Fertilizers and increased demands to store and dispose of industrial and municipal waste such as sewage has made this unsaturated zone to be contaminated(Thomas, 2007; Ukpaka 2013a–i; Amadi and Ukpaka, 2015, 2015a; Ikenyiri and Ukpaka, 2014, 2014a).

This zone is typically the first subsurface soil environment to encounter surface applied agrochemicals and industrial contaminants and hence all surface and subsurface chemical concentrations and subsequent environmental impact are inextricably linked to the Physical, Biological and Chemical dynamics within the zone; which include Sorption-Desorption, Volatilization, Photolysis, Degradation etc (Looney and Falta, 2000; Ukpaka and Otabiri, 2013; Ukpaka and Pele, 2012, Ukpaka and Amdai, 2014).

Unlike the aquifer of the saturated zone below, the unsaturated zone is not a source of readily available water for human consumption but it is of a great importance in providing water and nutrients that are vital to the biosphere, and intensively used for the cultivation of plans, construction of building and disposal of waste as earlier mentioned. The zone is majorly or often the main factor controlling water movement to the aquifer. Thus, it strongly affects the rate and management of groundwater (Ukpaka, 2015; Ukpaka and Ogoni, 2015; Theophilus et al., 2012; Prommer et al., 1999; Ruhdi and Mohammend, 2010; Daichao and David, 1999; Eluozo et al., 2012; Ewing et al., 1999). It is often regarded as a filter that removes undesired substances from groundwater. To some extent this is true but a more general fact is that the flow rate and chemical reactions in the unsaturated zone controls whether, where and how fast contaminants enters groundwater supplies (Looney and Falta, 2000). Understanding the physical, biological and

chemical processes of this zone is therefore crucial in determining the quality of ground water for human consumption.

Modeling the spread of pollutant in soil is complicated due to non-random spatial and temporal variations of physical, biological and chemical component of the soil environment (Stagnitti et al., 1995; Jack et al., 1998; Jacquas et al., 2008; Looney et al., 2000; Maddalera et al., 2007).

The two main flow mechanisms in this zone are the Matrix (uniform) flow and the Preferential (funnel, macro pores) flow mechanism. Preferential flow is the uneven and often rapid movement of contaminate through porous media, typically soil characterized by regions of enhanced flux that a small fraction of media such as worm holes, root holes, cracks etc., participates in most of the flow, allowing much faster transport of a range of contaminants through soil and create significant consequence for groundwater quality. While Matrix flow is a relatively slow an even movement of water and solute (contaminants) through the soil while sampling all pore spaces, obeying the convective-dispersive theory which assumes that water flow an average flow path through soil. Both types of flow occur often simultaneously but have considerably different consequence for water flow and chemical leaching (discharge, escape). The chemical composition of preferential flow reflects the concentration of water near the surface while matrix flow represents the concentration of water around the drain (Mckone and Bennatt, 2003; Mirbagheri, 2004; Muibat and Jimoh, 2013; Okoromah and Ukpaka, 2013; Parlenge and Hill, 1995; Witterson and Nicholson, 1996).

The spread of pollutant in soil is controlled by the flow of fluid (liquid or gaseous phase) in the soil environment. Model describing the spread of pollutants through the soil is discussed in most cases by the convective-dispersive equation also called the Richard's equation. The convective-dispersive equation is a combination of the diffusion and convection (advection) equation which describes Physical Phenomena where particles, energy or other physical quantities are transported inside a physical or chemical, and or biological system due to the following processes: Diffusion, Convection, Chemical reactions/degradation (Van and Alves, 1982; Ukpaka et al., 2015; Ukpaka et al., 2011; Ukpaka et al., 2012; Simunek et al., 2013, 2001).

Contaminants transported by water in the soil can follow a variety of paths. In the simplest case, a one-dimensional convection takes place which can be accompanied by dispersion, adsorption and chemical reaction and probably other factors. Description of contaminants transport in terms of average water velocity v, by the convective-dispersive equation remains the first and fundamental transport equation to be considered (Stagnitti, et al., 1995; Amadi and Ukpaka, 2013, 2014, 2015; Schmidt and Gier, 1990; Schwarz et al., 2009).

Fig. 1a: Soil samples obtained from the vicinity of the Asphalt plant Company, Enito 3, Ahoada west LGA, ready for analysis in the Chemical Sciences Research Laboratory of the Niger Delta University, Wilberforce Island, Bayelsa State
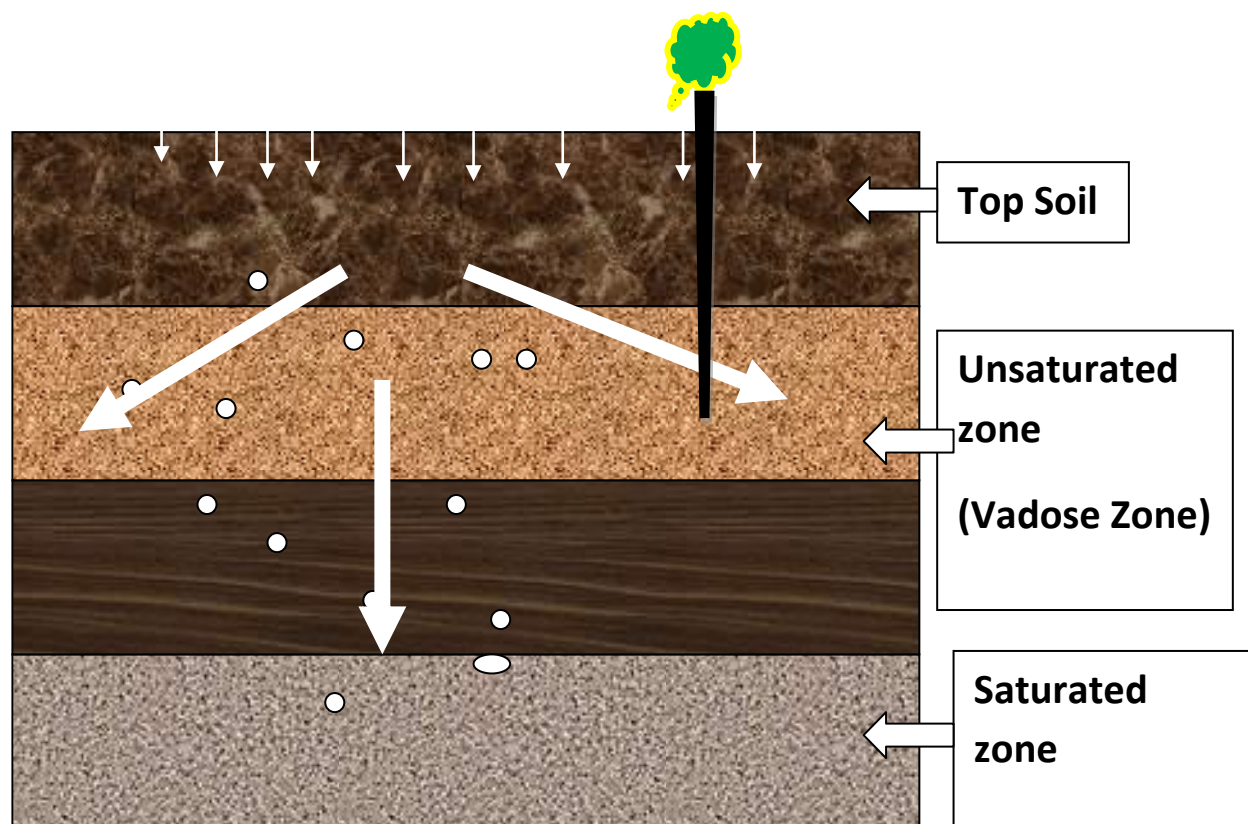
Fig. 1b: systematic distribution of contaminants in subsurface soil environment

Empirical modeling depends on the availability of representative data for model development and validation of already existing models. Also it seeks to approximate the unknown functional relationship by some mathematical functions based on experimentally gathered data from the physical system (Ukpaka, 2012; Singh et al., 2007). In this

Empirical or Data base modeling work, Experimental data are fitted to the developed mathematical model. The resulting correlation structure is then used to predict the behavior of the variables in the soil environment.

The following procedures apply to the proceedings of this project work. Development of Mathematical model, Collection of Experimental data from the analysis of the Physio-Chemical parameters of soil samples collected from within the vicinity of the H & H Asphalt plant Company in Enito 3 community in Ahoada West L.G.A., Specification of the correlation structure between variables (concentration & depth), Postulation of model, and Estimation of unknown parameters from the correlation structures, Validation of the model.

Soil pollution has a number of harmful effects on the Ecosystem and Humans, Plants and Animals (Schmidt and Gier, 1990; Parson et al., 1990; Phelps et al., 1990; Ukpaka, 2011a; Ukpaka and David, 2010).The harmful effects of soil pollution may come from direct contact with polluted soil or from contact with other related resources such as Water, food which has been grown on or came indirect contact with polluted soil. Soil doesn't need to be highly contaminated to be harmful to human. It may not significantly be polluted but still can be harmful to humans through bioaccumulation (the uptake of chemicals by a plant or animal, and the storage of the chemicals or its degradation product within the tissues of that organism), (Watson et al., 1998).

Soil pollution is a major Environmental Engineering problem on both Local and International level. One of the largest pollution problems in the world today is the threat of safe drinking water. The source of human drinking water is a mixture of surface water reservoirs and groundwateraquifer (Simunek, et al., 2003; Fathi and Emad, 2012). It's vitally important to the sustainability and safety of our society that this water source be carefully observed and protected from pollutants. A significant source of pollutant of drinking water supplies comes from industrial activities and discharge of untreated or under treated storm water runoff (Fathi and Emad, 2012). Industrial activities do not only involve discharge of pollutants onto the soil environment but also in some cases pave ways for the rapid flow of contaminants through the soil environment.

The degree of soil Pollution is therefore, to be assessed through the determination of these various parameters such as heavy metals, Petroleum Hydrocarbon as well as the Physio-chemical properties of the soil environment.

The following are the aims and objectives of this research work :to establish the impact of the industrial activities on the soil environment within the area of study ,to ascertain its impact on the vegetation, to monitor and predict its rate of deposition and diffusion on the soil environment, to ascertain the migration of the contaminant in due course on the underground water, examine the degree of pollution as a result of industrial activity within the operational region of the company under investigation and drawing of conclusion from my findings.

Scope of the Study includes: Mathematical model formulation - to develop a mathematical model that will monitor and predict the rate of pollutant deposition and diffusion in soil environment, the developed mathematical model will be related to the formulation of the Empirical model, the Empirical model developed will be related in terms of physico-chemical parameters of the soil environment within the area of this study; experimental analysis of Microbial, Hydrocarbon and of the Physio-chemical parameters of soil samples collected from within the vicinity of the H & H Asphalt Plant Company in Enito 3 Community, at Ahoada West L.G.A, River State, Nigeria; validation of the developed model using the Experimental data and testing of the model.

The study of the fate of chemical pollutant in soil environment is vital, for sustaining agricultural productivity and land utility which are directly related to human safety. It's imperative to know, that whatever degrades, damages, or destroy the soil environment ultimately have an impact on human life and may threaten our very ability to survive (Gupta and Shukle, 2006; Patil and Tijare, 2001; Singh and Mathur, 2005). Hence, an understanding of the cause, distribution processes and effects, will enable us to find a way to either stop or reduce this problem (Gupta and Deshpande, 2004).

The developed model will be used to monitor the impact of the pH and conductivity of the soil environment within the area of operation.

The model can be used to study the fate of contaminate in any soil environment with available data.

The developed model can be used to predict the quality of underground water supplies and refills.

MATERIALS AND METHODS

Sample collection

Soil samples were collected within the vicinity of the H & H Asphalt Plant Company Located at Enito 3, a village in Ahoada West Local Government Area, River State, Nigeria. Soil samples were obtained from the surface (top soil) to a depth of 60cm. A total of five (5) soil samples were collected within the interval of 15cm depth, top soil 0-0.5cm, 15cm, 30cm, 45cm and 60cm. The samples were transported to the Chemical Sciences Research Laboratory of the Niger Delta University for analysis, for the determination of the Physio-chemical Parameters.

pH measurement

10g of air-dried soil samples were weighed into 100ml beaker and 20ml of distilled water added. The slurry was stirred with glass rod to break up the ions until homogeneous slurry is obtained. The electrode of the pH meter was then dipped into the slurry and gently stirred. The electrode is then left to stand in the slurry until a steady pH reading is obtained.

Table 1: Results of experimental determination of physico-chemical parameters

Parameter	Unit	Depth (cm) (Dp)					DPR Limit/ Intervention level
		0.00-5.00	15.00	30.00	45.00	60.00	
pH		5.72	5.87	5.93	6.07	6.02	6.5-8.5
Conductivity	μS	666.00	033.00	055.00	091.00	101.00	

Table 2: Theoretical computation of pH values investigated

Depth (cm) (Dp)	pH (pH) $_{EV}$	Dp*pH	Dp^2	Dp^2*pH	Dp^3	Dp^4
0.00-5.00	5.72	0.00	0.00	0.00	0.00	0.00
15.00	5.88	88.20	225.00	1323.00	3375.00	50625.00
30.00	5.93	177.90	900.00	5337.00	27000.00	810000.00
45.00	6.07	273.15	2025.00	12291.75	91125.00	4100625.00
60.00	6.02	361.20	3600.00	21672.00	216000.00	12960000.00
$\sum$Dp =150.00	$\sum$pH =29.62	$\sum$Dp*pH =900.45	$\sum Dp^2$ =6750.00	$\sum Dp^2$*ph =40623.75	$\sum Dp^3$ =337500.00	$\sum Dp^4$ =17921250.00

Table 3: Comparison of experimental, theoretical and validated values for pH investigation

Depth(cm) (Dp)	Experimental pH values (pH) $_E$	Theoretical developed Model pH values (pH) $_{TDM}$	Validated model pH values (pH) $_{VM}$
00.00-5.00	5.72	5.72	5.77
15.00	5.88	5.88	5.84
30.00	5.93	5.98	5.92
45.00	6.07	6.05	5.99
60.00	6.02	6.06	6.07

Conductivity

Air-dried and sieved soil samples (10 g) were dissolved in 200ml of distilled water and allowed to stand for 30min. the slurry was then filtered. The conductivity of the filtrate was then measured. A steady read out from the conductivity meter, a Hanna Digital Conductivity meter was used.

Contaminants transport model in soil environment

The spread of pollutants in soil is controlled by the flow of fluid in the soil environment (Stagnitti et al 1995;Rushdi, and Mohammed, 2010; Mirbagheri, 2004; Fathiand Emad, 2012).

Model describing pollutant fate in the soil environment is discussed here by the convective-dispersive equation which describes the physical phenomena of mass transfer where particles are transported in a media due to convection, diffusion chemical reactions and biological transformation.
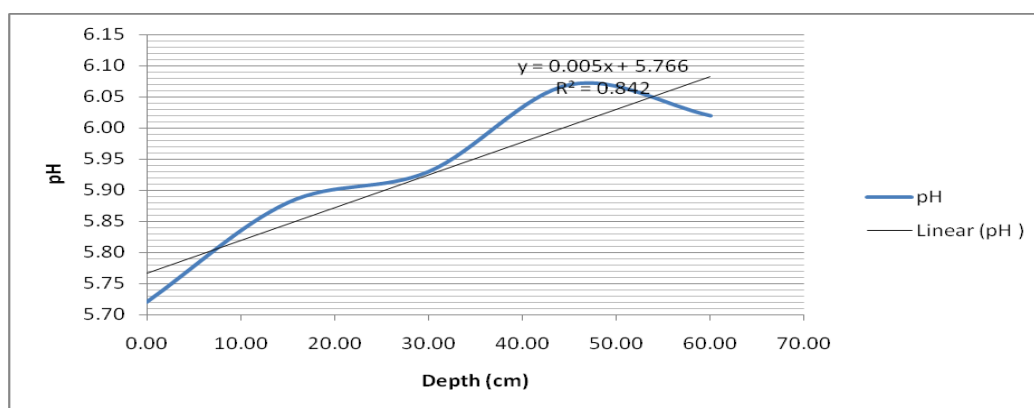
The convective-dispersive equation can be derived in a straight forward way from the continuity equation which states that the rate of change of a scalar quantity in a differential control volume is given by flow and diffusion into and out of the system along with any generation or consumption inside the control volume. The Figure 1 shows a control volume in a soil environment describing the behavior of contaminants in the soil environment.

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Table 4: Theoretical computation of conductivity values investigated

Depth (cm) (Dp)	Conductivity (µS) (CND)$_{EV}$	DP*CND	DP^2	DP^2*CND	DP^3	DP^4
0.00-5.00	666.00	0.00	0.00	0.00	0.00	0.00
15.00	33.00	495.00	225.00	7425.00	3375.00	50625.00
30.00	55.00	1650.00	900.00	49500.00	27000.00	810000.00
45.00	91.00	4095.00	2025.00	184275.00	91125.00	4100625.00
60.00	101.00	6060.00	3600.00	363600.00	216000.00	12960000.00
$\sum$Dp =150.00	$\sum$CND =946.00	$\sum$Dp*CND =12300.00	$\sum$Dp^2 =6750.00	$\sum$Dp^2*CND =604800.00	$\sum$Dp^3 =337500.00	$\sum$Dp^4 17921250.00

Table 5: Comparison of experimental, theoretical and validated values for conductivity investigation

Depth (cm) (Dp)	Conductivity (µS) (CND)$_{EV}$	Conductivity (µS) (CND)$_{TDM}$	Conductivity (µS) (CND)$_{VM}$
00.00-5.00	666	589.31	403.60
15.00	033	203.59	296.41
30.00	055	003.71	189.22
45.00	091	-010.32	082.03
60.00	101	161.51	-025.16

Fig. 2: Experimental pH values against depth

The model was developed by considering the illustration as shown in figure 2. Then, the concentration term can be expressed by the following differential equation. The material balance expression for the system is as shown in equation (1a), thus:

Contaminant accomulation rate = Diffusion − covection(advection) − adsorption rate − Irreversible decay   (1a)

Empirical model approach

The mathematical representation of equation (1a) is shown below

$$\frac{\partial C}{\partial t} = \mathbf{\nabla}.(D\mathbf{\nabla}C - vC) - R - f \tag{1b}$$

$$\frac{\partial C}{\partial t} = \mathbf{\nabla}.(D\mathbf{\nabla}C) - \mathbf{\nabla}.(vC) - \frac{\partial Ca}{\partial t} - f \tag{2}$$

Where, C is the concentration, v is the average water velocity, D is the dispersion coefficient (diffusivity), Ca is the concentration of the adsorbed chemical contaminant and f is the irreversible reaction decay rate.

By expanding the equation (2) into three coordinates

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right) - v\frac{\partial C}{\partial x} - \frac{\partial Ca}{\partial t} - f + \frac{\partial}{\partial y}\left(D\frac{\partial C}{\partial y}\right) - v\frac{\partial C}{\partial y} - \frac{\partial Ca}{\partial t} - f + \frac{\partial}{\partial z}\left(D\frac{\partial C}{\partial z}\right) - v\frac{\partial C}{\partial z} - \frac{\partial Ca}{\partial t} - f \tag{2b}$$

1.   Assuming one-dimensional flow. Then, the y- and z- coordinates are neglected and assumed to be zero, the equation becomes:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right) - v\frac{\partial C}{\partial x} - \frac{\partial Ca}{\partial t} - f \tag{3}$$

The equation (3) can be rearranged of as

$$\partial C/\partial t - \partial/\partial x\,(D\,\partial C/\partial x) + v\,\partial C/\partial x = -\partial Ca/\partial t - f \tag{4}$$

2.   Assuming that contaminant are transported at a steady state within the soil environment.

Then, equation (3) reduces to a second order deferential equation

$$-D\frac{d^2C}{dx^2} + v\frac{dC}{dx} + f = 0 \tag{5}$$

Since diffusion is taking place the negative sign is insignificant to the system.

$$D\frac{d^2C}{dx^2} + v\frac{dC}{dx} + f = 0 \tag{5}$$

Equation (5) is expressed as a quadratic equation of the form

DX^2+vX+f=0   (6)

Where $X^2 = \dfrac{d^2C}{dx^2}$ and $X = \dfrac{dC}{dx}$,The variable X is the depth.

The concentration is expressed as a function of depth

C (X) = DX^2+vX+f   (7)

Empirical modeling depends on the availability of representative data for model development and validation of already existing models. Its function includes approximation of unknown functional relationship by some mathematical functions based on the experimentally gather data from the physical system.

The resultant quadratic equation relates the Diffusivity (D), water Velocity (v) and the irreversible decay rate (f). Using the least square method the experimental data are fitted to the above equation (7) for all the parameters that were analyzed as:

C (X) = DX^2+vX+f   (8)
CND (X) = DX^2+vX+f   (9)

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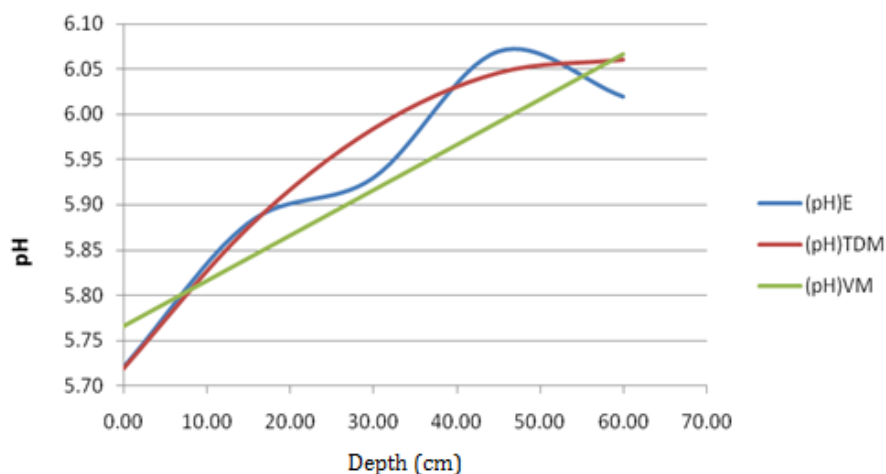
Fig. 3: Comparison of experimental, theoretical model and validated model pH (concentration) against depth

RESULTS AND DISCUSSION

The results obtained from the research work are presented in Tables and Figures for the various investigations conducted on the empirical model approach to the evaluation for the degree of pollutant deposition and diffusion in soil environment.

The result presented in figure 2 illustrates the relationship between the experimental pH values of contaminated soil environment and depth. Increase in pH values was observed with increase in soil depth. The variation in the pH values can be attributed to the variation in depth as well as the degree of deposition and diffusion of contaminants in the soil environment. The polynomial of the curve was established as Y = 0.005X + 5.766 and the square root of the best fit is given as R^2= 0.842. The expression above can be written as C^E_{pH}=0.005Dp+5.766 for the experimental data. While the theoretical equation developed is shown in Eq. 10.

$$C^T_{pH}= -0.000105Dp^2+0.012Dp+5.719 \qquad (10)$$

From figure 3, it is seen that the theoretical developed model pH values, experimental pH values and the validated pH values were plotted against soil depth. The relationship shows a good match, indicating the reliability of the developed model when compared with the experimental values. Points of interception were obtained at various regions such as at 8.0cm depth were the pH value is 5.80 for both theoretical validated and experimental pH values.

The figure 4 shows the soil experimental conductivity values plotted against depth. The conductivity of the top soil is higher and gradually reduces as the depth increases. The polynomial of the curve is displayed as $(COND)_{VM}$ =-7.146Dp+409.6 and the coefficient of determination is given as R^2=0.400. The theoretical

developed model for conductivity is given as $(COND)_{TDM}$= 0.413Dp^2-31.92Dp+589.31

Figure 5 illustrate the relationship between the Experimental, Theoretical model and Validated model Conductivity values against Depth. It is seen that there is a good match up to a depth of 50cm. The theoretical model values show a good relationship compared with the experimental values. Increase in conductivity was observed latter on for both experimental and the theoretical model.

CONCLUSIONS

From the comparison of the results shown there's a reasonable level of agreement between the experimented theoretical and validated model, indicating the viability of the developed model. The concentration of the parameters analyzed can be predicted and various depths, the coefficient of diffusivity (D), the water velocity (v) and the irreversible decay rate (f) can as well be obtained from the equation developed.

The acidic concentration decreases with increase in diffusion depth. This can be attributed to infiltration system taking place in the soil environment. The result obtained when compared with world health organization standard revealed variations on pH value within the area of 0.05 cm to 30.00 cm depth indicating the impact of contaminant as a result of diffusion. There is tendency that in a few years to come other depths of 35 cm and beyond will be affected generating stronger acidic nature of the soil.

From the result shown the conductivity at the surface is very high, which means there's no significant diffusion at the subsurface of the soil environment. However, in due time there's the tendency of migration of contaminant into the soil thereby increasing the conductivity. Bioaccumulation is a major factor, it's really invisible and if not properly track can cause serious environmental challenge, health issues etc.
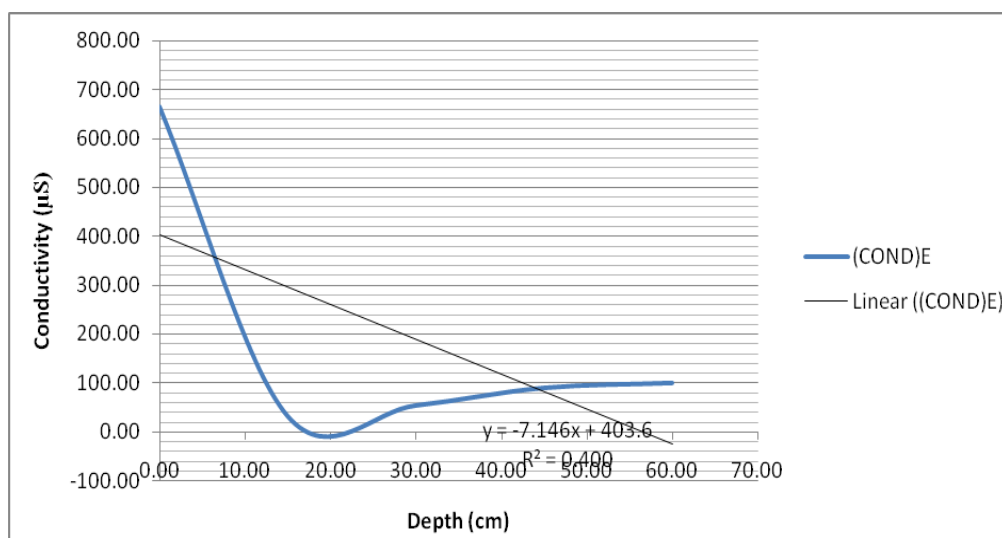
Fig. 4: Experimental conductivity values against depth
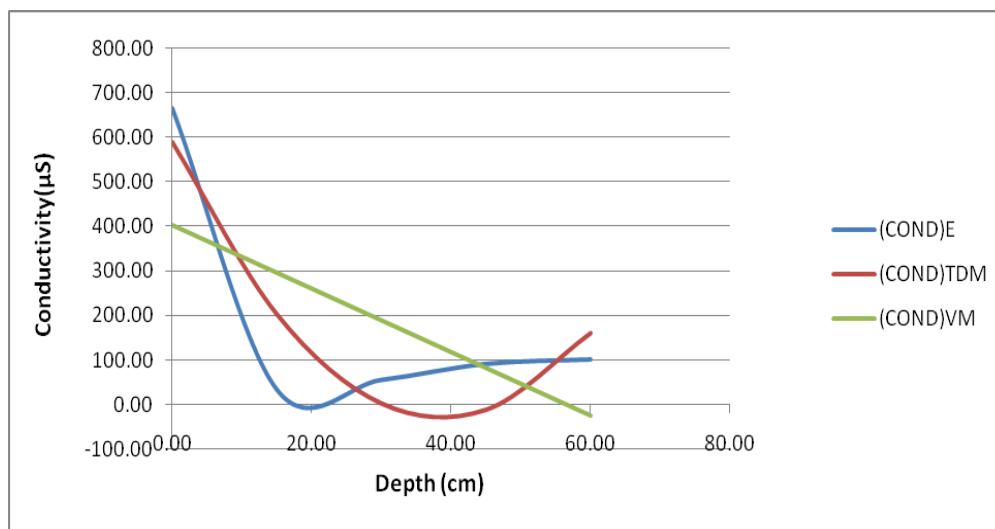
Fig. 5: Comparison of experimental, theoretical model and validated model Conductivity values against depth

This research work can be adopted or carried out quarterly to check the trend at which pollutants have infiltrated into the soil environment and surrounding water bodies to avoid bioaccumulation. Bioaccumulation was the result of the Minamata Mercury poison (1956) which resulted to very diversifying issues that affected Humans, Animals, Plants and the Company Chisso Corporation, as wellafter paying for damage which cost them billions of dollars.

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